Claims

[c1] 1.A pressure vessel for processing at least one material in a supercritical fluid, the pressure vessel comprising:

a)a capsule for containing said at least one material and said supercritical fluid in a substantially air-free environment, said capsule being self-pressurizing;

b)a pressure transmission medium for maintaining an outer pressure on said capsule, said pressure transmission medium surrounding said capsule;

c)a heating system for heating said capsule, said heating system comprising at least one heating element insertable in said pressure transmission medium such that said at least one heating element is proximate to said capsule and a wattage control system electrically coupled to said at least one heating element, wherein said wattage control system provides power to said at least one heating element;

d)a restraint to contain and hold in place said capsule, said pressure transmission medium, and said at least one heating element, wherein said restraint maintains said capsule, said pressure transmission medium, and said at least one heating element at a constant pressure; and

e)at least one seal for preventing escape of said pressure transmission medium, said at least one seal being disposed between said restraint and said pressure transmission medium.

[c2] 2.The pressure vessel of Claim 1, wherein said heating system further comprises at least one temperature sensor disposed proximate to said capsule for measuring a temperature of said capsule.

[c3] 3.The pressure vessel of Claim 2, wherein said at least one temperature sensor comprises at least one of a thermocouple, a thermistor, and an optical fiber coupled to an optical pyrometer.

[c4] 4.The pressure vessel of Claim 2, wherein said wattage control system provides closed loop temperature control in response to at least one signal generated by said at least one temperature sensor.

[c5] 5.The pressure vessel of Claim 1, wherein said at least one heating element is an electrically resistant heating element comprising at least one of at least one

foil, at least one tube, at least one ribbon, at least one bar, and at least one wire, and combinations thereof.

[c6] 6. The pressure vessel of Claim 1, wherein said at least one heating element comprises at least one of graphite, nichrome, niobium, titanium, tantalum, stainless steel, nickel, chromium, zirconium, molybdenum, tungsten, rhenium, hafnium, platinum, silicon carbide, and combinations thereof.

[c7] 7.The pressure vessel of Claim 1, wherein said heating system differentially heats a first portion of said capsule to a first temperature and a second portion of said capsule to a second temperature.

[c8] 8.The pressure vessel of Claim 1, further including a clamp for loading at least one portion of said restraint and reducing at least one of a longitudinal stress and an axial stress on at least one portion of said restraint.

[c9] 9.The pressure vessel of Claim 8, further including at least one gasket disposed between said clamp and at least one portion of said restraint.

[c10] 10.The pressure vessel of Claim 9, wherein said at least one gasket includes an electrically insulating gasket, wherein said electrically insulating gasket is formed from at least one of natural rubber, synthetic rubber, polyester film, polyimide, fluorocarbon polymer, tetrafluoroethylene fluorocarbons, fluorinated ethylene-propylene, pyrophyllite, talc, olivine, magnesium oxide, calcium carbonate, calcium oxide, strontium oxide, barium oxide, textilite, a glued paper composite, merylinite clay, bentonite clay, sodium silicate, and hexagonal boron nitride.

[c11] 11.The pressure vessel of Claim 10, wherein said at least one gasket includes an electrically conductive element within said electrically insulating gasket, wherein said electrically conductive element is formed from at least one of molybdenum, graphite, tungsten, tantalum, niobium, nickel, nickel alloy, iron, iron alloy, and combinations thereof.

[c12] 12.The pressure vessel of Claim 9, wherein said at least one gasket includes an electrically conductive gasket, wherein said electrically conductive gasket is

formed from at least one of copper, brass, molybdenum, graphite, nickel, cobalt, iron, and stainless steel.

[c13] 13.The pressure vessel of Claim 1, wherein said at least one seal comprises a top seal and a bottom seal.

[c14] 14.The pressure vessel of Claim 13, wherein said top seal comprises a top end cap and said bottom seal comprises a bottom end cap, and wherein said top end cap and said bottom end cap are formed from steel.

[c15] 15.The pressure vessel of Claim 14, wherein said top end cap further includes a deformable ring to provide a seal between said clamp and said restraint.

[c16] 16.The pressure vessel of Claim 1, wherein said pressure transmission medium is thermally stable up to about 1000 ° C and has an internal friction of less than about 0.2.

[c17] 17.The pressure vessel of Claim 16, wherein said pressure transmission medium is a solid up to about 1300 ° C.

[c18] 18.The pressure vessel of Claim 16, wherein said pressure transmission medium comprises at least one of an alkali metal halide, talc, pyrophyllite, molybdenum disulfide, graphite, hexagonal boron nitride, silver chloride, calcium fluoride, strontium fluoride, calcium carbonate, magnesium oxide, zirconium oxide, merylinite clay, bentonite clay, and sodium silicate.

[c19] 19.The pressure vessel of Claim 18, wherein said pressure transmission medium comprises at least one of sodium chloride, sodium bromide, and sodium fluoride.

[c20] 20.The pressure vessel of Claim 1, wherein said restraint comprises at least one die, at least one punch, and a press.

[c21] 21.The pressure vessel of Claim 20, wherein said at least one die is one of a straight-wall die, an angled-wall die, and a concave-wall die.

[c22] 22.The pressure vessel of Claim 20, wherein said at least one die is formed from one of cemented tungsten carbide and hardened steel.

[c23] 23.The pressure vessel of Claim 20, wherein said at least one die is contained within at least one steel compression ring.

[c24] 24.The pressure vessel of Claim 20, further including a cooling sleeve disposed between said at least one die and said at least one compression ring, wherein said cooling sleeve includes at least one cooling channel for circulating a cooling medium therethrough.

[c25] 25.The pressure vessel of Claim 20, wherein said at least one die is contained within at least one of at least one tension-wound steel wire and at least one steel ribbon.

[c26] 26.The pressure vessel of Claim 20, wherein at least one of said at least one punch is a flat-bottomed punch, and wherein said flat-bottomed punch is squeezed against said at least one die by said press.

[c27] 27.The pressure vessel of Claim 20, wherein said pressure vessel has a pressure response of less than about 0.2.

[c28] 28.The pressure vessel of Claim 27, wherein said pressure response is less than about 0.05.

[c29] 29.The pressure vessel of Claim 1, wherein said restraint comprises a multi-anvil press.

[c30] 30.The pressure vessel of Claim 29, wherein said multi-anvil press comprises at least four anvils.

[c31] 31.The pressure vessel of Claim 29, wherein said multi-anvil press comprises at least four pistons.

[c32] 32.The pressure vessel of Claim 30, further comprising a plurality of support plates, wherein each of said plurality of support plates is disposed between of said at least four anvils and said pressure transmission medium.

[c33] 33.The pressure vessel of Claim 29, wherein said multi-anvil press has a pressure response of less than about 0.2.

[c34] 34.The pressure vessel of Claim 33, wherein said pressure response is less than about 0.05.

[c35] 35.The pressure vessel of Claim 1, wherein said restraint comprises at least two end flanges, a die disposed between said at least two end flanges, and at least one fastener joining said at least two end flanges.

[c36] 36.The pressure vessel of Claim 35, wherein each of said at least two end flanges further includes a structural support for reinforcing each of said at least two end flanges.

[c37] 37.The pressure vessel of Claim 35, wherein said structural support comprises an I-beam.

[c38] 38.The pressure vessel of Claim 35, wherein said at least one fastener comprises at least one of a bolt and a threaded rod.

[c39] 39.The pressure vessel of Claim 1, wherein said capsule is self-pressurizable from about 1 bar up to about 80 kbar.

[c40] 40.The pressure vessel of Claim 39, wherein said capsule is self-pressurizable up to between about 5 kbar and about 80 kbar.

[c41] 41.The pressure vessel of Claim 39, wherein said capsule is self-pressurizable up to between about 5 kbar and about 60 kbar.

[c42] 42.A capsule for containing at least one material and a supercritical fluid in a substantially air-free environment, wherein said capsule has at least one wall, a closed end, and a sealed end defining a chamber therein for containing said at least one material and said supercritical fluid, and wherein said capsule is self-pressurizing.

[c43] 43.The capsule of Claim 42, wherein said capsule is formed from a malleable metal, and wherein said capsule has a low hydrogen permeability.

[c44] 44.The capsule of Claim 42, wherein said capsule is formed from a first material comprising at least one of stainless steel, copper, silver, gold, and platinum.

[c45] 45.The capsule of Claim 42, wherein said capsule includes an inert liner inserted into said chamber, wherein said inert liner is formed from a second material comprising at least one of gold, platinum, rhodium, palladium, silver, iridium, ruthenium, silica, and wherein said inert liner is between about 1 micron and about 5 mm thick, and wherein said second material is different from said first material.

[c46] 46.The capsule of Claim 42, wherein said at least one wall, said closed end, and said sealed end each have a thickness of between about 0.5 mm and about 25 mm.

[c47] 47.The capsule of Claim 42, wherein said capsule is self-pressurizable from about 1 bar up to about 80 kbar.

[c48] 48.The capsule of Claim 47, wherein said capsule is self-pressurizable up to between about 5 kbar and about 80 kbar.

[c49] 49.The capsule of Claim 47, wherein said capsule is self-pressurizable up to between about 5 kbar and about 60 kbar.

[c50] 50.A pressure vessel for processing at least one material in an a supercritical fluid, the pressure vessel comprising:

a)a capsule for containing said at least one material and said supercritical fluid in a substantially air-free environment, wherein said capsule has at least one wall, a closed end, and a sealed end defining a chamber therein for containing said at least one material and said supercritical fluid, and wherein said capsule is self-pressurizing;

b)a pressure transmission medium for maintaining an outer pressure on said capsule, said pressure transmission medium surrounding said capsule;

c)a heating system for heating said capsule, said heating system comprising at least one heating element insertable in said pressure transmission medium such that said at least one heating element is proximate to said capsule, at least one temperature sensor disposed proximate to said capsule for measuring a temperature of said capsule, and a wattage control system electrically connected to said at least one heating element and said at least one

temperature sensor, wherein said wattage control system provides power to said at least one heating element and controls said temperature;

d)a restraint to contain and hold in place said capsule, said pressure transmission medium, and said at least one heating element, wherein said restraint maintains said capsule, said pressure transmission medium, and said at least one heating element at a constant pressure; and

e)at least one seal for preventing escape of said pressure transmission medium, said at least one seal being disposed between said restraint and said pressure transmission medium.

[c51] 51.The pressure vessel of Claim 50, wherein said at least one heating element is an electrically resistant heating element comprising one of at least one foil, at least one tube, at least one ribbon, at least one bar, and at least one wire, and combinations thereof.

[c52] 52. The pressure vessel of Claim 50, wherein said at least one heating element comprises one of graphite, nichrome, niobium, titanium, tantalum, stainless steel, nickel, chromium, zirconium, molybdenum, tungsten, rhenium, hafnium, platinum, silicon carbide, and combinations thereof.

[c53] 53.The pressure vessel of Claim 50, wherein said at least one temperature sensor comprises at least one of a thermocouple, a thermistor, and an optical fiber coupled to an optical pyrometer.

[c54] 54.The pressure vessel of Claim 50, wherein said wattage control system provides closed loop temperature control in response to at least one signal generated by said at least one temperature sensor.

[c55] 55.The pressure vessel of Claim 50, wherein said heating system differentially heats a first portion of said capsule to a first temperature and a second portion of said capsule to a second temperature.

[c56] 56.The pressure vessel of Claim 50, further including a clamp for loading said restraint and reducing at least one of a longitudinal stress and an axial stress on at least one portion of said restraint.

[c57] 57.The pressure vessel of Claim 56, further including at least one gasket disposed between said clamp and at least one portion of said restraint.

[c58] 58.The pressure vessel of Claim 57, wherein said at least one gasket includes an electrically insulating gasket, wherein said electrically insulating gasket is formed from at least one of natural rubber, synthetic rubber, polyester film, polyimide, fluorocarbon polymer, tetrafluoroethylene fluorocarbons, fluorinated ethylene-propylene, pyrophyllite, talc, olivine, magnesium oxide, calcium carbonate, calcium oxide, strontium oxide, barium oxide, textilite, a glued paper composite, merylinite clay, bentonite clay, sodium silicate, and hexagonal boron nitride.

[c59] 59.The pressure vessel of Claim 58, wherein said at least one gasket includes an electrically conductive element within said electrically insulating gasket, wherein said electrically conductive element is formed from at least one of molybdenum, graphite, tungsten, tantalum, niobium, nickel, nickel alloy, iron, iron alloy, and combinations thereof.

[c60] 60.The pressure vessel of Claim 59, wherein said at least one gasket includes an electrically conductive gasket, wherein said electrically conductive gasket is formed from at least one of copper, brass, nickel, cobalt, iron, and stainless steel.

[c61] 61.The pressure vessel of Claim 50, wherein said at least one seal comprises a top seal and a bottom seal.

[c62] 62.The pressure vessel of Claim 61, wherein said top seal comprises a top end cap and said bottom seal comprises a bottom end cap, and wherein said top end cap and said bottom end cap are formed from steel.

[c63] 63.The pressure vessel of Claim 62, wherein said top end cap further includes a deformable ring to provide a seal between said clamp and said restraint.

[c64] 64.The pressure vessel of Claim 50, wherein said pressure transmission medium is thermally stable up to about 1000 ° C and has an internal friction of less than about 0.2.

[c65] 65.The pressure vessel of Claim 64, wherein said pressure transmission medium is a solid up to about 1300 ° C.

[c66] 66.The pressure vessel of Claim 64, wherein said pressure transmission medium comprises at least one of an alkali metal halide, talc, pyrophyllite, molybdenum disulfide, graphite, hexagonal boron nitride, silver chloride, calcium fluoride, strontium fluoride, calcium carbonate, magnesium oxide, zirconium oxide, merylinite clay, bentonite clay, and sodium silicate.

[c67] 67.The pressure vessel of Claim 66, wherein said pressure transmission medium comprises at least one of sodium chloride, sodium bromide, and sodium fluoride.

[c68] 68.The pressure vessel of Claim 50, wherein said restraint comprises at least one die, at least one punch, and a press.

[c69] 69.The pressure vessel of Claim 68, wherein said at least one die is one of a straight-wall die, an angled-wall die, and a concave-wall die.

[c70] 70.The pressure vessel of Claim 68, wherein said at least one die is formed from one of cemented tungsten carbide and hardened steel.

[c71] 71.The pressure vessel of Claim 70, wherein said at least one die is contained within at least one steel compression ring.

[c72] 72.The pressure vessel of Claim 70, further including a cooling sleeve disposed between said at least one die and said at least one compression ring, wherein said cooling sleeve includes at least one cooling channel for circulating a cooling medium therethrough.

[c73] 73.The pressure vessel of Claim 70, wherein said at least one die is contained within at least one of at least one tension-wound steel wire and at least one steel ribbon.

[c74] 74.The pressure vessel of Claim 68, wherein at least one of said at least one punch is a flat-bottomed punch, and wherein said flat-bottomed punch is squeezed against said at least one die by said press.

[c75] 75.The pressure vessel of Claim 74, wherein said pressure vessel has a pressure response of less than about 0.2.

[c76] 76.The pressure vessel of Claim 75, wherein said pressure response is less than about 0.05.

[c77] 77.The pressure vessel of Claim 50, wherein said restraint comprises a multi-anvil press.

[c78] 78.The pressure vessel of Claim 77, wherein said multi-anvil press comprises at least four anvils.

[c79] 79.The pressure vessel of Claim 77, wherein said multi-anvil press comprises at least four pistons.

[c80] 80.The pressure vessel of Claim 78, further comprising a plurality of support plates, wherein each of said plurality of support plates is disposed between of said at least four anvils and said pressure transmission medium.

[c81] 81.The pressure vessel of Claim 77, wherein said multi-anvil press has a pressure response of less than about 0.2.

[c82] 82.The pressure vessel of Claim 81, wherein said pressure response is less than about 0.05.

[c83] 83.The pressure vessel of Claim 50, wherein said restraint comprises at least two end flanges, a die disposed between said at least two end flanges, and at least one fastener joining said at least two end flanges.

[c84] 84.The pressure vessel of Claim 83, wherein each of said at least two end flanges further includes a structural support for reinforcing each of said at least two end flanges.

[c85] 85.The pressure vessel of Claim 83, wherein said structural support comprises an I-beam.

[c86] 86.The pressure vessel of Claim 83, wherein said at least one fastener comprises at least one of a bolt and a threaded rod.

[c87] 87.The pressure vessel of Claim 50, wherein said capsule is formed from a malleable metal, and wherein said capsule has a low hydrogen permeability.

[c88] 88.The pressure vessel of Claim 50, wherein said capsule is formed from a first material comprising at least one of stainless steel, copper, silver, gold, and platinum.

[c89] 89.The pressure vessel of Claim 50, wherein said capsule further includes an inert liner inserted into said chamber, wherein said inert liner is formed from a second material comprising at least one of gold, platinum, rhodium, palladium, silver, iridium, ruthenium, silica, and wherein said inert liner is between about 1 micron and about 5 mm thick, and wherein said second material is different from first material.

[c90] 90.The pressure vessel of Claim 50, wherein said at least one wall, said closed end, and said sealed end each have a thickness of between about 0.5 mm and about 25 mm.

[c91] 91.The pressure vessel of Claim 50, wherein said capsule is self-pressurizable from about 1 bar up to about 80 kbar.

[c92] 92.The pressure vessel of Claim 91, wherein said capsule is self-pressurizable up to between about 5 kbar and about 80 kbar.

[c93] 93.The pressure vessel of Claim 91, wherein said capsule is self-pressurizable up to between about 5 kbar and about 60 kbar.

[c94] 94.A method of using a pressure vessel to process at least one material at high temperature and high pressure in the presence of a supercritical fluid, the method comprising the steps of:

a)providing a sealed capsule containing the at least one material and a solvent that forms a supercritical fluid, wherein the capsule is self-pressurizing;

b)providing a pressure vessel comprising a restraint for containing the sealed capsule, a pressure transmission medium disposed within the pressure vessel, and at least one heating element disposed within the pressure transmission medium and electrically coupled to a wattage control system;

c)disposing the sealed capsule within the pressure transmission medium such that the sealed capsule is proximate to the at least one heating element;
d)placing the pressure vessel containing the pressure transmission medium, the sealed capsule, and the at least one heating element in a press;
e)pressurizing the press to apply a predetermined pressure to the pressure vessel, the pressure transmission medium, the sealed capsule, and the at least one heating element;
f)providing electrical power from the wattage control system to the at least one heating element, thereby heating the sealed capsule to a predetermined temperature, wherein the solvent contained within the sealed capsule becomes a supercritical fluid and wherein the supercritical fluid generates a predetermined pressure within the sealed capsule; and
g)counterbalancing the predetermined pressure within the sealed capsule by maintaining an equivalent pressure with the restraint and transmitting the equivalent pressure through the pressure transmission medium, wherein the at least one material is processed at high temperature and high pressure in the presence of a supercritical fluid.

[c95] 95.The method of Claim 94, wherein the restraint comprises at least one die, at least one punch, and a hydraulic press, and wherein the pressure transmission medium and the heating element are disposed within the die; and wherein the step of disposing the sealed capsule within the pressure vessel comprises disposing the sealed capsule within the die such that the sealed capsule is proximate to the at least one heating element.

[c96] 96.The method of Claim 95, wherein the restraint comprises a die, a top punch, and a bottom punch, wherein the top punch and the bottom punch oppose each other, and wherein the step of pressurizing the press to apply a predetermined pressure to the pressure vessel comprises pressurizing the press to apply a predetermined pressure onto the top punch and the bottom punch.

[c97] 97.The method of Claim 96, wherein the step of pressurizing the press to apply a predetermined pressure to the pressure vessel comprises the steps of:
a)pressurizing the press to apply a first predetermined pressure onto the top

punch and the bottom punch; and

b)adjusting the predetermined pressure to maintain the top and bottom punches at one of a fixed position and a stroke as the capsule is heated and pressure builds up therein.

[c98] 98.The method of Claim 96, further including the step of inserting at least one temperature sensor within the pressure vessel such that the temperature sensor is disposed proximate to the sealed capsule, wherein the at least one temperature sensor is electrically coupled to the wattage control system.

[c99] 99.The method of Claim 98, wherein the step of providing electrical power from the wattage control system to the at least one heating element further comprises providing closed loop temperature control in response to at least one signal generated by the at least one temperature sensor.

[c100] 100.The method of Claim 99, wherein the step of providing closed loop temperature control in response to at least one signal generated by the at least one temperature sensor comprises:

a)providing closed loop temperature control in response to a first signal generated by a first temperature sensor disposed proximate to a first portion of the sealed capsule; and

b)providing closed loop temperature control in response to a second signal generated by second temperature sensor disposed proximate to a second portion of the sealed capsule.

[c101] 101.A method of processing at least one material at high temperature and high pressure in the presence of a supercritical fluid, the method comprising the steps of:

a)providing a sealed capsule containing the at least one material and a solvent that forms a supercritical fluid, wherein the capsule is self-pressurizing;

b)providing a pressure vessel comprising a restraint, a pressure transmission medium disposed within the restraint, and at least one heating element disposed within the restraint;

c)disposing the sealed capsule within the pressure transmission medium such that the sealed capsule is proximate to the at least one heating element;

d)heating the sealed capsule to a predetermined temperature by providing electrical power to the at least one heating element, wherein the solvent contained within the sealed capsule becomes a supercritical fluid and wherein the supercritical fluid generates a predetermined pressure within the sealed capsule; and

e)counterbalancing the predetermined pressure within the sealed capsule by applying a pressure to the restraint, wherein the at least one material reacts with the supercritical fluid within the sealed capsule.

[c102] 102.A metal nitride single crystal, wherein the metal nitride single crystal is formed by: enclosing a metal nitride source material and a solvent within a sealed capsule that is self-pressurizing; disposing the sealed capsule within a pressure vessel comprising a restraint, a pressure transmission medium disposed within the restraint, and at least one heating element disposed within the restraint; heating the sealed capsule to a predetermined temperature, wherein the solvent contained within the sealed capsule becomes a supercritical fluid and generates a predetermined pressure within the sealed capsule; and counterbalancing the predetermined pressure within the sealed capsule by applying a pressure to the restraint; wherein the metal nitride source material reacts with the supercritical fluid within the sealed capsule to form a metal nitride single crystal at high temperature and high pressure.

[c103] 103.The method of Claim 102, wherein said metal nitride comprises aluminum nitride.